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Summary of Doctoral Dissertation

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Degree requested Doctor of Life Science　　Applicant's name　　MUHAMMAD ILYAS

学 位 論 文 題 名
Title of Doctoral Dissertation

Synthesis of Large Area Bilayer-hybrid Gels and Exploration of Novel Functions
（大面積二分子膜ハイブリッドゲルの合成とその新機能探索）

Creation of novel hydrogels with a flexible, dynamic hierarchical structure in similar to biological systems, and discovery of its unprecedented functions are among the most fundamental but challenging topics in the research of soft matter. Amphiphilic molecules, which have both hydrophilic head group and hydrophobic tail, self-assemble in aqueous media into various hierarchical structures. Inducing such hierarchical structure composed of amphiphilic molecules into amorphous hydrogels is a promising approach to meet the above-mentioned challenges. Recently, macro-scale bilayer hybrid hydrogels (PDGI/PAAm), based on amphiphilic polymer PDGI poly(dodecyl glyceryl itaconate), have been developed. The gels exhibited excellent functions, such as structure color, high mechanical toughness, and stimuli responsive properties. This promises great potential of the bilayer-based hydrogels for diversified application. Developing large area bilayer-hybrid hydrogels with the supramolecular self-assembled structure is indispensable for these potential applications. This thesis aims to establish a method to fabricate high quality bilayer-based hydrogels thin films and explore novel functions of bilayer-hybrid gels.

The thesis consists of 8 chapters. In **Chapter 1**, a general introduction is shown. **Chapter 2** enlightens the background study.

In **Chapter 3**, the new method of synthesizing large area bilayer-hybrid gels and optimized experimental conditions are explored. It is concluded that, to produce large area, of several tens of cm scale, well oriented stable bilayers in hydrogels, controlling the incubation temperature of the precursor solution and its shear flow to the reaction cell prior to gelation are important. This finding is useful for the mass production of functional hydrogels regardless dimensions and exploration of novel functions.

In **Chapter 4**, the detailed investigation of the effect of water content, temperature, solvent, configuration of monomers, on the structure and properties of bilayer-hybrid gels are explored. It is concluded that by slowly reducing water content the layered microstructure is unperturbed, and ductile-brittle transition of the PAAm gel layers occurs at much higher water content (58 wt%) as compared to the water content (26 wt%) of bulk PAAm hydrogels. At this transition point, the thickness of PAAm gel layers is 52 nm, much higher than the reported literature of water confined at several nanometer length scale. Accompanied with such a transition, the free water in the lamellar gels disappeared. These results indicate the long-range ordering of water in the lamellar hydrogels, which is observed for the first time.

In co-solvent, the adaptive mechanical behavior of gels at various concentration is studied. Drying water from co-solvent produced air stable, soft, tough and stretchable ion gel based on bilayers. The hybrid gel synthesized by adding small amount of ionic liquid that perturb the structure of water shows self-healing. The ion gel exhibited an ultra-fast self-healing behavior due to multiple reversible interactions.

The hybrid-gels synthesized using amphiphilic racemic and chiral monomers significantly altered the packing of bilayers that showed different yielding criteria and deformation under mechanical test.

In **Chapter 5**, the effect of different types of drying and functions related to drying are explored. It is found that by different drying, the gel shows rainbow like patterning, uniform color stimulation, nacre mimetic iridescence metallic luster. Based on these results, beetle cuticle mimetic water-based rewritable photonic paper, micro patterning with butterfly wing mimetic iridescence, and jelly fish mimetic photoluminescence, are developed. These findings might open new way to create functional devices for various application.

In **Chapter 6**, the solvent free fabrication of soft elastomers based on amphiphilic PDGI polymer is performed. As a result, the elastomer shows malleability by roll pressing, high birefringence, high stiffness and toughness. The elastomer is also useful to understand various properties of rigid bilayers.

In **Chapter 7**, the glucose-responsive photonic hydrogels is developed. To induce the glucose responsibility, poly(3-acrylamidopheylboronic acid) is induced in the photonic PDGI/PAAm gel. The obtained hydrogel reversibly changes color in response to glucose and small deformation. The photonic hydrogel has potential application as non-invasive visual glucose sensor.

In **Chapter 8**, the concluding remarks are included based on the overall work.